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Report No. 1

DISSEMINATION AND AEROSOL BEHAVIOR OF HYDROPHOBIC POWDERS

Quarterly Report

by

D. E. BLAKE
R. C. CREWDSON
C. E. LAPPLE
R. A. SCHMIDT

January 1968

STATEMENT [illegible]

DEPARTMENT OF THE ARMY
EDGEWOOD ARSENAL
Research Laboratories
Physical Research Laboratory
Edgewood Arsenal, Maryland 21010

Contract DAAA15-68-C-0099

STANFORD RESEARCH INSTITUTE
Menlo Park, California

27

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DEPARTMENT OF THE ARMY
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Contract DAAA15-68-C-0099
Task 1B522301A08101

STANFORD RESEARCH INSTITUTE
Menlo Park, California

[redacted]

FOREWORD

The work described in this report was authorized under Task 1B522301A08101, Dissemination Investigations of Liquid and Solid Agents (U). The work was started in October 1967 and completed in January 1968.

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SUMMARY

A detailed research plan was established after review and evaluation of current data for simple chemical compounds at high pressure and high temperatures. Apparatus has been designed and is being constructed to recover particles produced by a shock and to minimize the probability of reagglomeration or further collisions. It is hoped that the apparatus can permit a permanent record of the particle distribution if this should be desirable.

Theoretical considerations indicate that giving aerosol particles a net unipolar charge will increase the rate at which an aerosol cloud issuing from an apparatus will expand for reasonable aerosol concentration and charge levels. The prime uncertainty rests with the difficulty of achieving the desired charge levels. A laboratory nozzle has been designed and fabricated to permit direct assessment of this concept, using corona for charging.

The project on the dissemination of hydrophobic powders has been divided into two tasks. Task 1 is a study of the behavior of solids subjected to shock loading and is being conducted by R. A. Schmidt and R. C. Crewdson. Task 2 is an investigation to determine whether electrically charged particles of CS agent can be used to improve the effectiveness of the dissemination process. Most of the work on Task 2 during the past quarter has been conducted by D. E. Blake and C. E. Lapple.

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DISSEMINATION AND AEROSOL BEHAVIOR OF HYDROPHOBIC POWDERS

Task 1 - Behavior of Solids Under Shock Loading

A. Aims and Objectives

The objective of this task is to determine the behavior of solids and compacted powders under conditions of shock loading. It is necessary to understand the response of materials to explosive shocking in order to accomplish the objective of the program, which is to optimize the particle size distribution of aerosols produced by explosive dissemination.

An immediate aim is to verify and extend conclusions arrived at in Project PAU-4900. To this end, experimental methods are being perfected for recovery of particles for study of size distributions and separating effects due to extraneous causes (such as collisions with container walls). The same technique will be used in further studies of brittle materials and compacted materials.

Unique experimental facilities at Edgewood Arsenal will be employed to verify conclusions reached in other parts of the program and to test ideas designed to lead more directly to a useful process.

B. Summary of Results During Quarter

After review and evaluation of current data for simple chemical compounds at high pressures and high temperatures,[1-11] a detailed research plan was established. This is discussed below for each phase of this task.

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An uneven effort has been necessary to carry out this program because of the time involved in preparing and planning the new types of experiments discussed below. Preparation and planning has consumed about 10 percent of the project funds with the remainder to be used in the next reporting period.

It is desirable to recover the particles produced by shock without allowing them to reagglomerate or to suffer further collisions in order to investigate further previous conclusions concerning specimen confinement and to accurately assess the efficiency of comminution in plane shock experiments. An apparatus designed to accomplish this is being constructed. It is a container that will catch the particles in flight in a liner, which subsequently may be either dissolved or hardened to produce a permanent record of the particle distribution.

1. Phase I

The initial experimental effort under Phase I of Task 1 will include investigations of variations in the characteristics of model solid compounds, matrix materials, and shock strength as stated in Proposal No. FGU-FRU-67-135 and in Contract No. DAAA15-68-0099.

a. Characteristics of model solid compounds. The shock comminution behavior of pressed powder samples of mixtures of simple chemical compounds will be examined under varying shock pressures. Simple compounds were chosen for examination to determine the influence of elastic parameters and physico-chemical properties on comminution efficiency. These data are needed to predict the response of stimulants and agents to shock.

Table I summarizes characteristics of compounds to be used in this work. MgO was chosen as a reference material because of its high shock impedance and because of the availability of data on its response to elastic waves at high temperatures and high pressures.[5]

Table I

CHARACTERISTICS OF MATRIX MATERIALS

Materials	Mol. Wt (g)	Mean At Wt (m/p)	Density (gm cm^{-3})	Volume per Ion Pair ($2M/p_\rho$)	Compress (Mb^{-1})	Shock Impedance (bar sec/cm)	Ratio Shock Impedance W/MgO	Ratio $2M/p_\rho$ W/MgO
Al_2O_3	101.9	20.5	3.5	11.7	0.38	2.68	1.3	1.04
NaBr	102.9	51.5	3.2	32.2	0.05	0.82	2.5	0.29
KBr	119.0	59.5	2.75	43.3	0.03	0.52	4.0	0.38
NH_4Cl	53.5	10.7	1.53	14.0	0.02	0.5	4.0	0.13
SiO_2	60.1	20.0	2.65	15.1	0.27	0.37	5.6	0.14
Teflon	100.0	16.7	2.16	15.4	0.01	0.29	7.1	0.13
Polyethylene	28.1	4.7	1.05	8.9	0.01	0.15	13.7	0.08
MgO	40.3	20.2	3.58	11.3	0.59	2.07	--	--

Separate MgO samples will be prepared as standards for the examination of comminution efficiency at different shock pressures. Since MgO powder requires a binder in the fabrication of pressed samples, it is planned to vary the binder materials. This will provide for study of the effects of binder materials on the efficiency of comminution of the powdered samples.

Data obtained from MgO experiments will provide the basis for interpretation of results from binary mixtures of MgO with the other compounds listed in Table I. A series of samples will be prepared by mixing MgO as an internal standard with different proportions of each material. The substances to be mixed with MgO were chosen to cover a range of shock impedances, compressibility, and volume per ion pair. The influence of shock impedance on initial agent breakup and dissemination was discussed by Poppoff.[9] Compressibility effects on materials experiencing shock was described by Doran and Linde.[10] The relationship of volume per ion pair to bulk modulus and chemical composition and structure was described by Anderson and Nafe.[3] The latter paper notes that the bulk modulus-volume relationship of oxides differs from that of other compounds. The planned samples are designed also to examine the possible influence of this fact on dissemination, because mixtures of oxides, halides and oxides, and organic compounds and oxides are included. Therefore the series of samples will provide for (1) comparison of matrix response to shock with respect to the standard and (2) evaluation of the degree of control on dissemination exerted by variations in material properties among samples. These results and those from (b) below will guide the preparation of samples so as to simulate agent CS more closely.

b. Hugoniot data. It is planned to conduct explosive shots at the SRI Corral Hollow Test Site (when this facility is completed in January 1968) to determine the Hugoniot relationships of CS and resorcinol. These data are desired to better define materials for prediction of response

to shock loading and to verify the validity of resorcinol as a simulant for CS so that it can be used in gas gun work. Instrumentation for these shots is in preparation. Edgewood Arsenal was requested to provide four 2-inch-diameter by 1/4-inch-thick samples each of resorcinol and CS for these experiments.

2. Phase II

Selected samples of MgO plus binder and mixtures of MgO with materials of varying shock impedance are in preparation for tests by the government. These experiments are designed to augment the plane shock work at Stanford Research Institute. Additional samples to examine other variables will be prepared as the program progresses.

C. Work Proposed During Next Quarter

In the next reporting period it is planned to accomplish the following:

1. Hugoniot shots on CS and resorcinol at 15, 50, 100, and 150 kilobar pressure, using explosives at the SRI Corral Hollow Test Site.
2. Shock wave dissemination and recovery shots of pressed powders of MgO and MgO mixtures using the SRI light gas gun facility.
3. Completion of samples for Edgewood aerosol tests.

Task 2 - Electrical Charging of Agent Particles

A. Objective

The objective of Task 2 is to determine if the effectiveness (spreading of cloud and deposition from cloud) of CS aerosol clouds can be enhanced by electrical charging of the agent particulates during the dissemination process. The primary goal will be to generate a net unipolar-charged cloud on a sufficiently large scale to demonstrate that cloud expansion can be enhanced and surface deposition increased by the presence of the charge. A secondary goal will be to evaluate various systems for achieving a net unipolar charge in a cloud in order to develop or permit selection of an optimum system for field applications with various logistic limitations.

B. Summary of Results During Quarter

Calculations based on available information have indicated that significant cloud spreading can be achieved by particle charge levels that should be attainable by corona charging. There is little reason to question the validity of the theoretical predictions of spreading provided the desired particle charge levels can be obtained. The desired particle charge levels have been obtained with corona charging in the past. However, such applications have dealt with much lower particle concentrations (and hence lower currents). We believe that any failure to achieve predicted performance will probably be due to a failure to achieve the necessary charge levels with the high particle concentrations also required to achieve the desired degree of cloud spreading.

For this reason, a laboratory-scale system has been designed to rapidly check such potential problems. This laboratory equipment is

currently being assembled. In this system it is proposed to jet a 1-inch-diameter aerosol stream into free air at velocities of 10 to 100 ft/sec and agent simulant rates of 0.1 to 1 lb/min. A corona charging process will be used and the degree of cloud spreading will be observed under various operating conditions.

C. Work Proposed During Next Quarter

The performance of the laboratory equipment will be determined. If the equipment gives the expected cloud spreading, designs for a field test unit will be made and submitted to Edgewood Arsenal. If the desired spreading is not attained, attempts will be made to establish the cause of failure and develop any necessary modifications. It should be noted that the laboratory-scale unit is expected to have a capacity corresponding to the lowest capacity at which a field test unit is expected to operate.

D. Theory

A given cloud of unipolar-charged particles will expand because of the repulsive force between particles. This cloud expansion results in a dilution of the cloud.

A previous report[9] on a related project gives the equations governing the behavior of such clouds. For a cloud of unipolar particles, the rate at which the spatial concentration of particles changes due to electrostatic repulsion between particles is given by

$$
\begin{aligned}
-(d\,\ln c_p/dt) &= -(d\,\ln n_p/dt) \\
&= 2k_{C}\epsilon\delta_v c_p \mathcal{E}^2_{ps}/\mu\rho_p = 1/t_{sd}
\end{aligned}
\qquad (1)
$$

This rate of concentration change is an intrinsic property of the aerosol, depending on neither size nor shape of cloud. Equation 1 would hold at any point within the cloud.

For a cloud of unipolar and uniformly distributed charged particles bounded by a surface equidistant from a center of symmetry [either a plane (flat volume); a line (circular cylindrical volume); or a point (spherical volume)], the electrostatic field intensity at the surface of the cloud is given by

$$\mathcal{E} = (6c_p\mathcal{E}_{ps}/\rho_p D_p)(V/A)$$

$$= 6m\mathcal{E}_{ps}/A\rho_p D_p \tag{2}$$

and the rate of radial expansion of the surface of the cloud is given by

$$u_R = (V/A)/t_{sd} = (2k_C \epsilon \delta_v c_p \mathcal{E}^2_{ps}/\mu\rho_p)(V/A) \tag{3}$$

The above describes the behavior of a given cloud after it is formed. Conditions at any point in space within this cloud will change with time. If instead of such a given original cloud, particles are continuously added to a region in space, a cloud of given properties will form as the result of the quasi-equilibrium attained when the rate of particle addition to that point in space is the same as the rate at which particles leave. Conditions at any point in space within this cloud will not change with time. This quasi-equilibrium condition is attained when

$$c_p = 4w_p/\pi D^2 u \tag{4}$$

in the case of a rod-shaped cloud.

Also at any cross section of this rod-shaped quasi-equilibrium cloud,

$$(V/A) = D/4 \quad . \tag{5}$$

Thus, for long circular cylindrical clouds, Equations 2 and 3 may be written as

$$(\mathcal{E}/\mathcal{E}_{ps}) = (3/2\ \rho_p D_p)(c_p D) \tag{6}$$

and

$$(u_R/u) = (k_C \varepsilon \delta_v / 2\mu\rho_p)(c_p D\ \mathcal{E}^2_{ps}/u) \tag{7}$$

respectively. By use of Equations 4 and 6, Equation 7 may be written in the following alternative forms:

$$(u_R/u) = (2k_C \varepsilon \delta_v / \pi\mu\rho_p)(w_p \mathcal{E}^2_{ps}/Du^2) \tag{8a}$$

$$(u_R/u) = (\pi k_C \varepsilon \delta_v \rho_p D_p^2/18\mu)(D\mathcal{E}^2/w_p) \tag{8b}$$

$$(u_R/u) = (k_C \varepsilon \delta_v D_p/3\mu)(\mathcal{E}\mathcal{E}_{ps}/u) \tag{8c}$$

The derivation of Equation 8 tacitly assumed a constant value of c_p throughout the cloud volume. Consequently, these equations will only be valid if the cloud is not being diluted. In practice this would mean that Equation 8 is valid only for small values of (u_R/u).

The ratio (u_R/u) is a measure of divergence of the gas jet constituting the aerosol cloud due to electrostatic expansion of the cloud. Any divergence due to physical entrainment of ambient gas by the jet will be in addition to any electrostatic expansion.

Figures 1, 2, and 3 give numerical values of (u_R/u) calculated from Equations 8a, b, and c, respectively, for the specific parameter values indicated in each figure. The fluid property values for each figure correspond to those for air at 1 atm, 25°C.

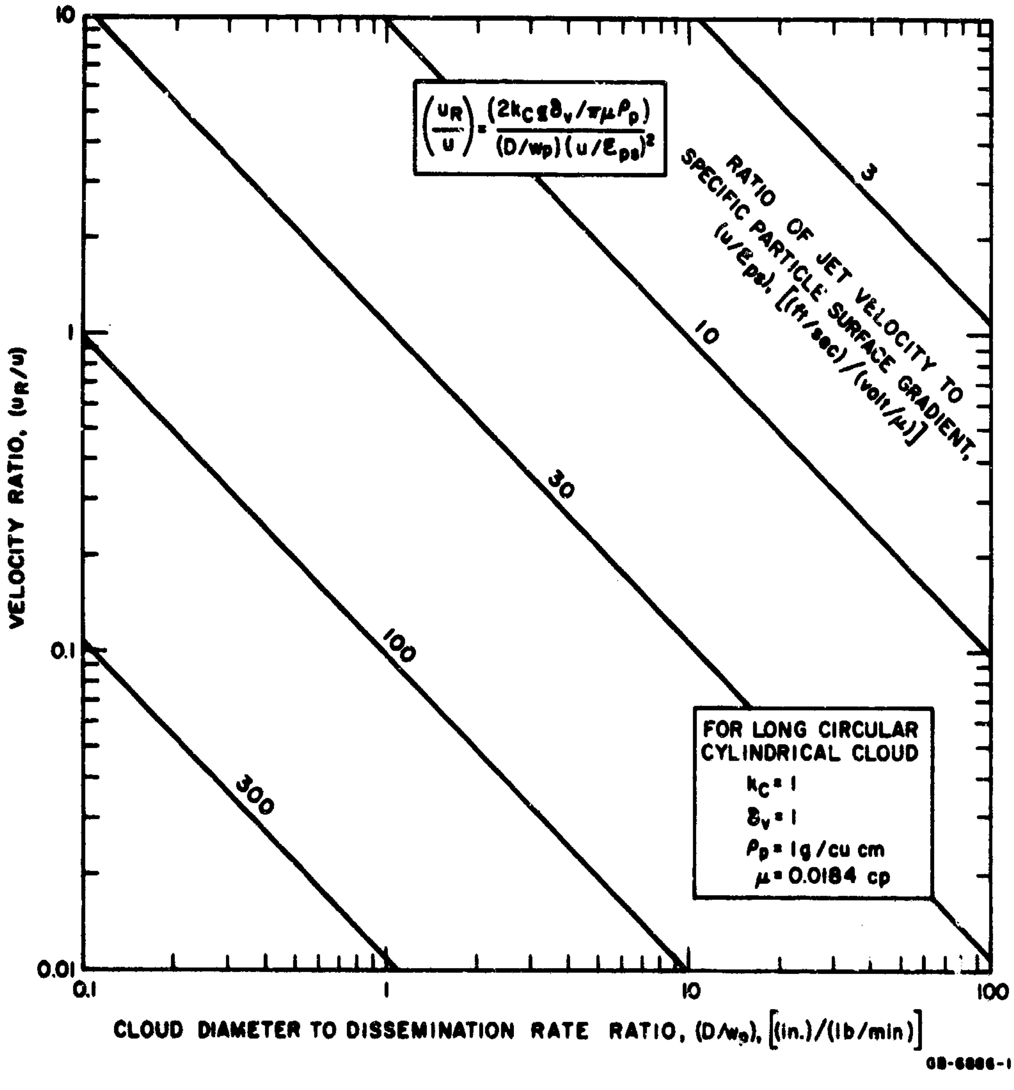

FIG. 1 CLOUD EXPANSION AS FUNCTION OF CLOUD DIAMETER, DISSEMINATION RATE, CLOUD JET VELOCITY, AND PARTICLE CHARGE

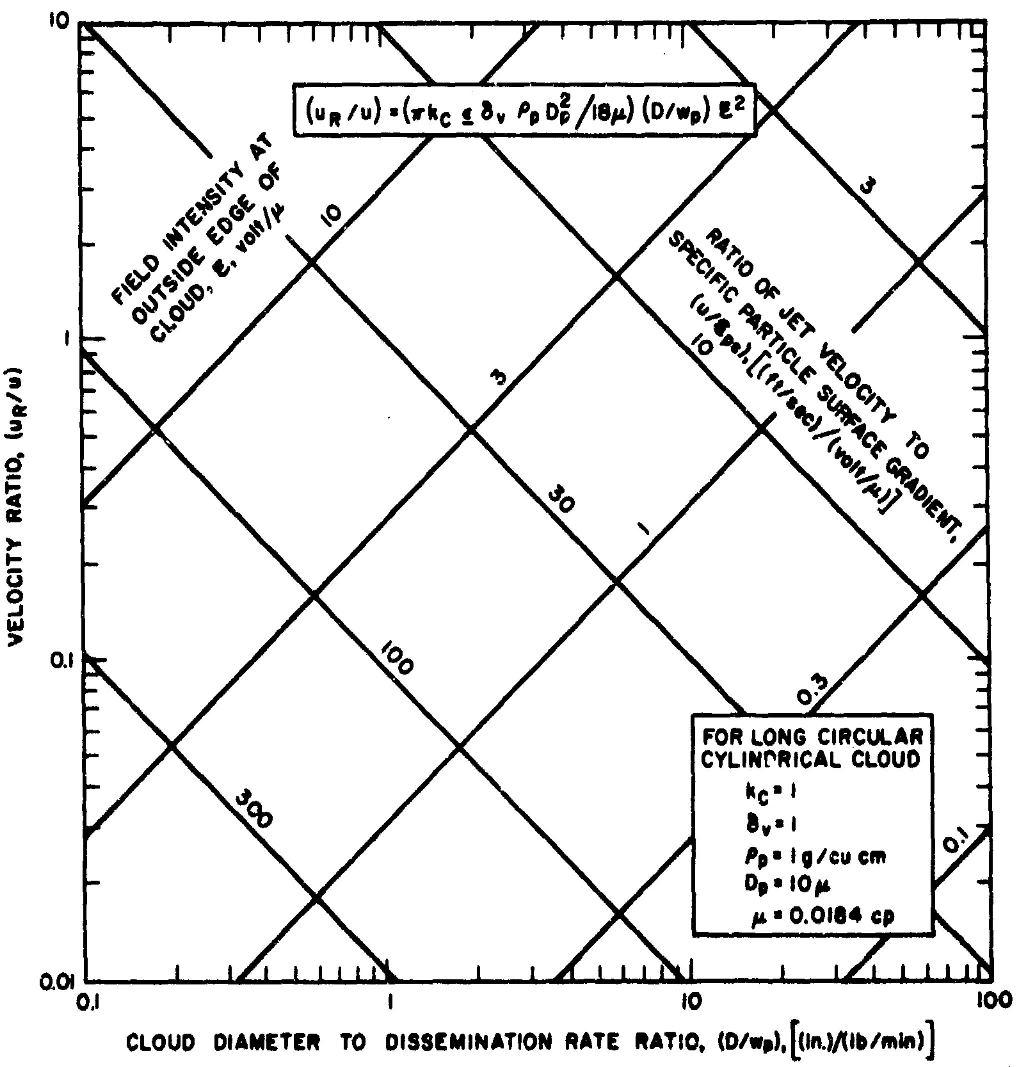

FIG. 2 CLOUD EXPANSION AS FUNCTION OF CLOUD DIAMETER, DISSEMINATION RATE, AND OUTER CLOUD FIELD INTENSITY

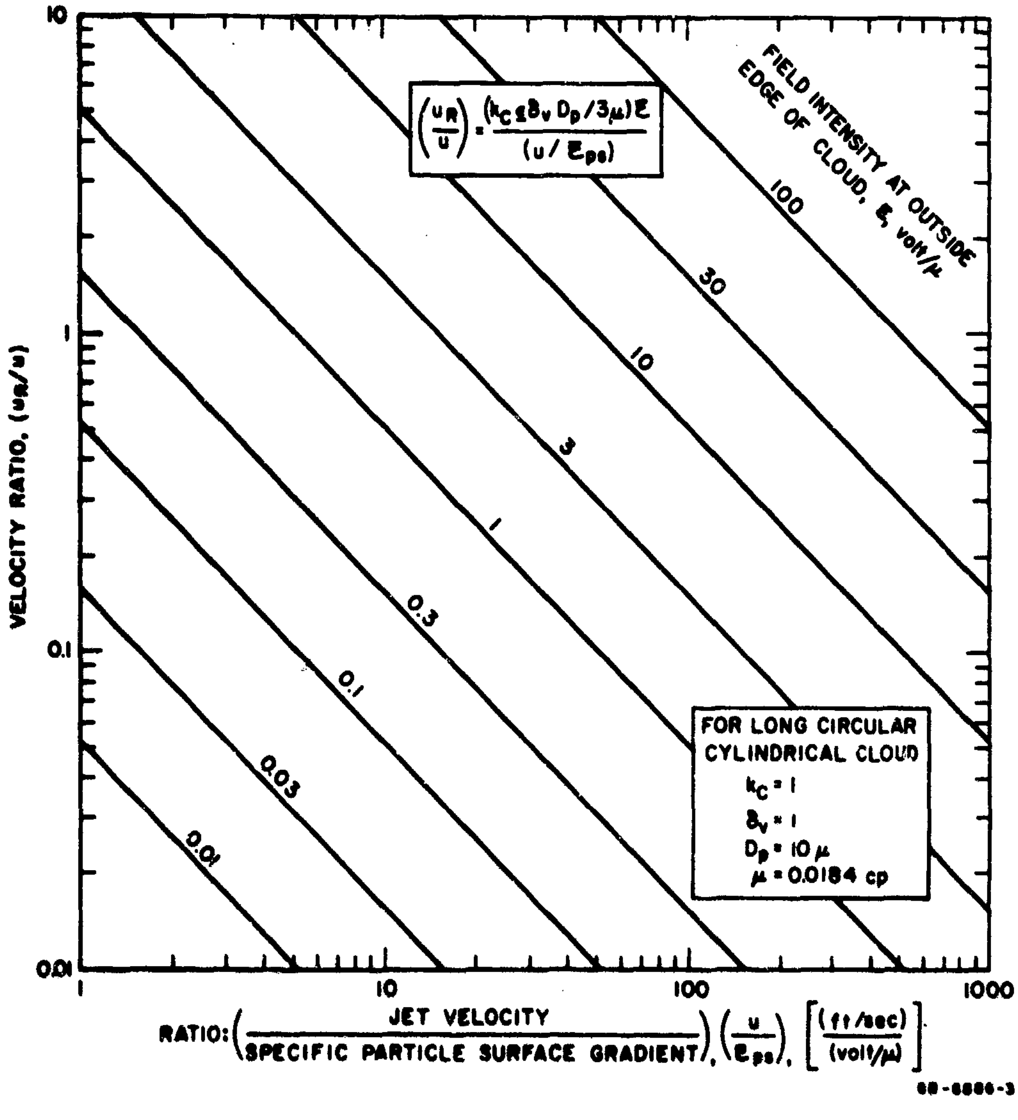

FIG. 3 CLOUD EXPANSION AS FUNCTION OF CLOUD JET VELOCITY, PARTICLE CHARGE, AND OUTER CLOUD FIELD INTENSITY

In the above equations, all the particles are assumed to have the same size and charge, with charge being expressed in terms of $\mathcal{E}_{ps}$. The theoretical current needed to provide the specified charge level is given by

$$I_p = (6\underline{\epsilon}\delta_v/\rho_p D_p)(w_p \mathcal{E}_{ps}) \tag{9}$$

Figure 4 gives representative values of I for the specific conditions cited.

E. Discussion

In the above theoretical development, the ratio (u_R/u) is actually a measure of the cloud expansion and would correspond to the tangent of the half angle of the expanding conical cloud. The theoretical development is based on a highly simplified model, in which cloud jet velocities are assumed uniform at any cross section and which is limited to small expansion rates [i.e., small values of (u_R/u)]. In practice, the jet will expand due to dilution with surrounding air even when the particles are uncharged. This expansion usually corresponds to a cone angle of approximately 10 to 14°. Thus, an uncharged cloud would have an apparent (u_R/u) value of 0.09 to 0.12 (tan 5° to tan 7°). Any expansion due to homopolar charged particles would be superimposed on this. Also, the velocity profile in the jet will become markedly nonuniform due to cloud dilution.

Despite these complications, the simple relationships given in Figs. 1, 2, and 3 should serve as a basis for bracketing those operating conditions at which electrostatic cloud expansion should be observable. Thus, to distinguish from expansion due to ordinary cloud dilution, it is necessary to provide conditions for which the value of (u_R/u) due to particle charge is significant or large compared to the apparent value due to dilution (0.1).

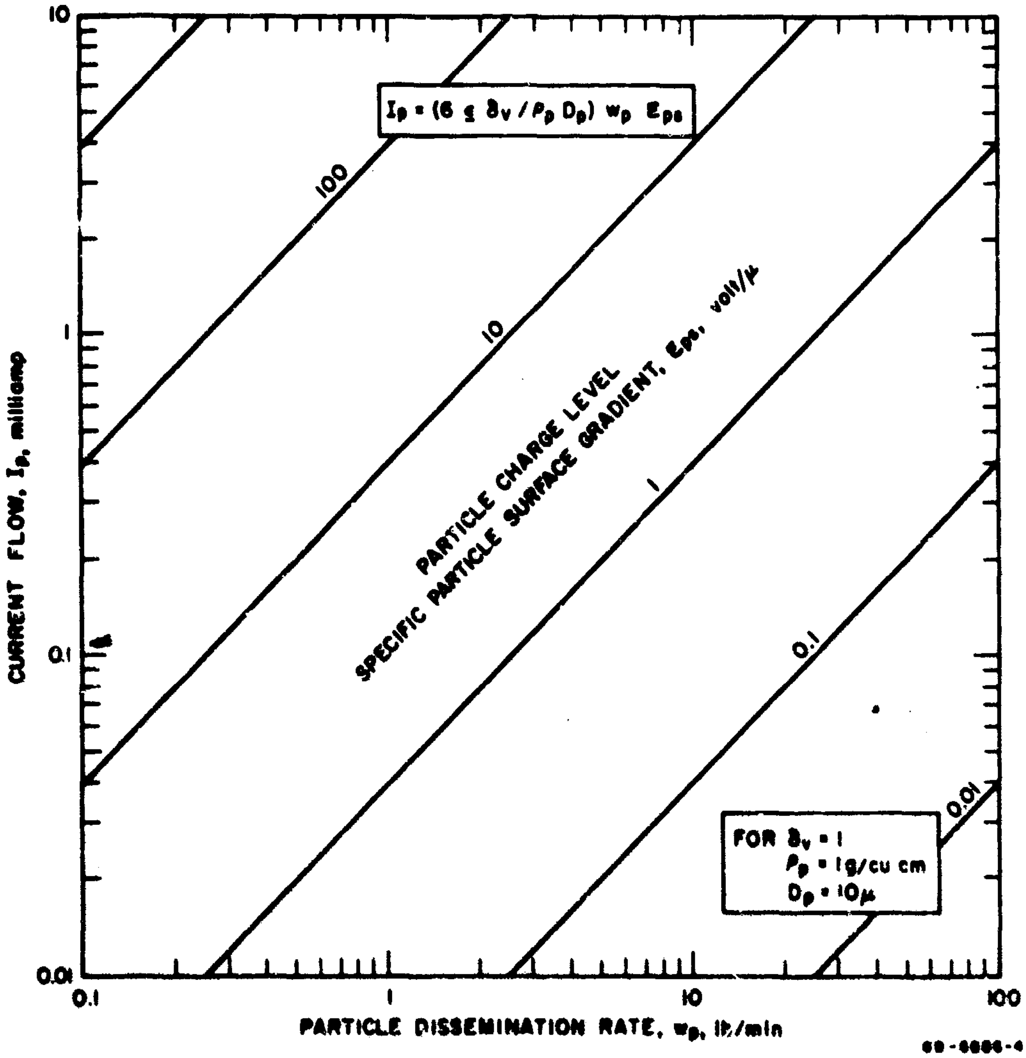

FIG. 4 THEORETICAL CURRENT REQUIRED FOR VARIOUS CHARGE LEVELS AND DISSEMINATION RATES

For purposes of designing demonstration conditions, we may assume the following:

1. Particle charge levels ($\mathcal{E}_{ps}$) of 1 v/μ. These are levels which have been achieved by means of corona charging.

2. Outer cloud charge levels ($\mathcal{E}$) of 3 v/μ. This is the level at which the outer edge of the cloud would be expected to go into corona. While it is not known that this will be an effective limitation, we must assume so for present purposes.

The demonstration design problem, therefore, becomes one of selecting reasonable and attainable conditions at which a value of (u_R/u) of 0.1 or greater is to be expected with the above assumptions.

For conditions normally encountered, it is apparent from Fig. 3 (or Equation 8) that cloud jet velocities must be less than 160 ft/sec if the ratio (u_R/u) is to be greater than 0.1 for 10 μ-diameter particles. For 1-μ-diameter particles, jet velocities must be less than 16 ft/sec. These velocity limitations apply regardless of the size of the cloud or solids rates (subject to assumptions above).

Arbitrarily selecting a 1-inch-diameter jet, the following corresponding values can be predicted from Fig. 3 (or Equation 8c) for the case of 10-μ particles and particle charge levels of 1 v/μ.

Solids rate, lb/min:	0.1	0.3	0.5	1	1
Gas velocity, u, ft/sec:	10	50	50	50	100
Gas flow rate, ft^3/min	3.3	16	16	16	33
Solids concentration, g/liter	0.5	0.3	0.5	1.0	0.5
Expansion ratio, u_R/u	1.0	0.12	0.20	0.40	0.10
Outer cloud field intensity, $\mathcal{E}$, v/μ	1.9	1.1	1.9	3.7	1.9

Thus, a 1-inch cloud jet would appear to be a reasonable arrangement that should be capable of demonstrating cloud expansion, provided that the assumed average net unipolar particle charge level can be attained.

Although the model above is considerably simpler than the actual situation can be, it should serve as a guide for designing experiments aimed at verifying theoretical predictions. At a later date, we will attempt to solve cases of more complex and realistic models.

F. Experimental Equipment

On the basis of the foregoing, the equipment shown in Fig. 5 has been designed and constructed. Powder will be fed into a compressed air jet at an annular section (1/2-inch OD, 1/4-inch ID). The mixture will then be gradually decelerated to a 1-inch ID circular cross section. Just prior to discharging into the atmosphere, the aerosol will be exposed to a corona discharge.

The disseminated cloud will all be collected in a bag filter, where the bag becomes part of the inner insulated electrode of a condenser. This inner electrode is then grounded to the outer electrode through an ammeter that is used to determine the total net charge of a given sign on the particles. The charging current will be measured by an ammeter in the high potential circuit. If there is no loss of charging current, the two ammeters should read the same.

The behavior of the cloud will be observed, both with and without electrical charging, under various operating conditions. If significant differences are noted, quantitative measurements of cloud expansion will be made. Runs will last about 30 to 60 sec.

It is the purpose of these tests to indicate semiquantitatively that the predicted performance can be achieved in fact.

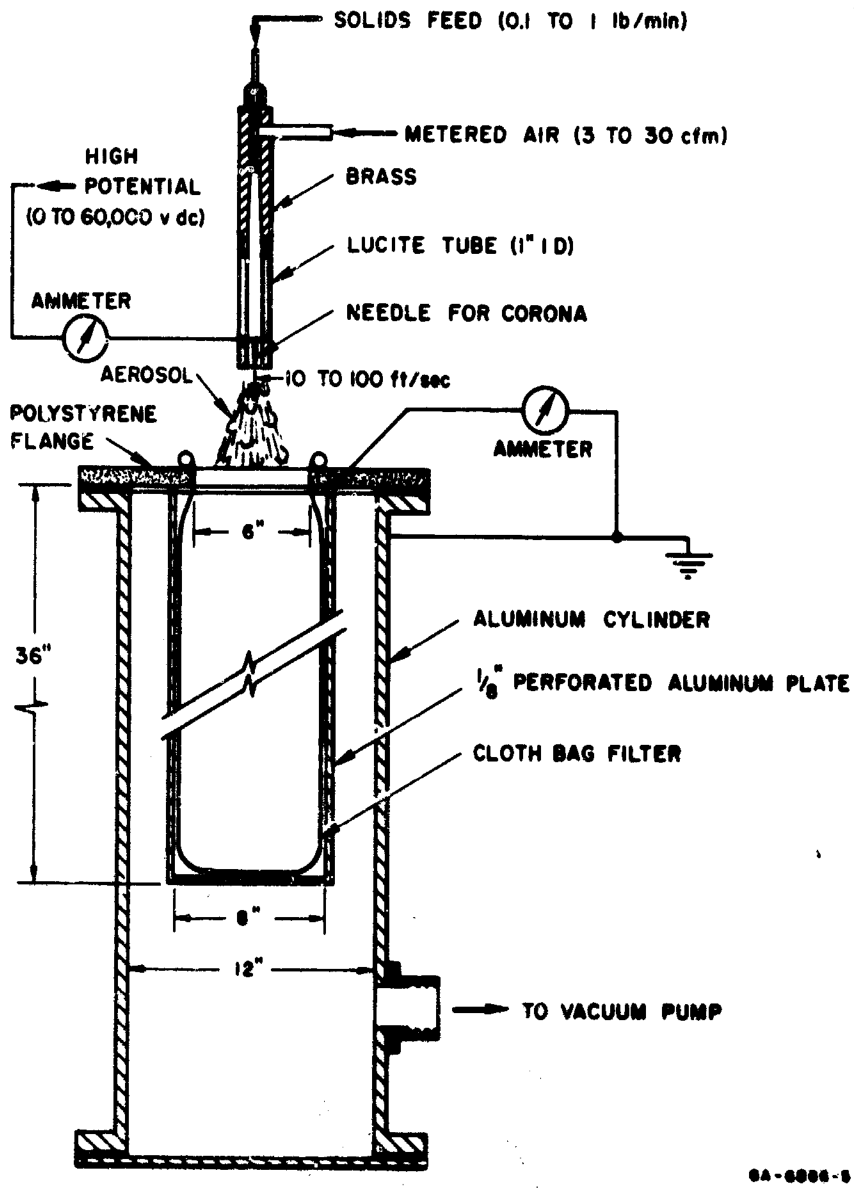

FIG. 5 EXPERIMENTAL EQUIPMENT

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NOMENCLATURE

(GLOSSARY)

A = surface area of a bounded volume V, m^2

A_f = area in cloud normal to gas flow direction, m^2

c_p = mass concentration of particles, kg/m^3

D = diameter of cylindrical cloud, m

D_p = particle diameter, m

$\mathcal{E}$ = potential gradient or field intensity at outer e ,e of cloud, v/m

$\mathcal{E}_{ps}$ = specific particle surface gradient for particle of size D_p,
$= \mathcal{Q}_p/\pi\underline{\epsilon}\delta D_p^2$, v/m

I_p = particle charging current, amp

k_C = Stokes-Cunningham correction factor, dimensionless

m = total mass of particles, kg

n_p = number concentration of particles, particles/m^3

$\mathcal{Q}_p$ = charge on particle of diameter D_p, coulomb

t = time, sec

t_{sd} = specific dilution time = $\mu\rho_p/2k_C\underline{\epsilon}\delta_v c_p \mathcal{E}^2_{ps}$, sec

u = average gas velocity through area A_f, m/sec

u_R = radial outward velocity of expanding cloud, m/sec

V = cloud volume, m^3

w_p = agent dissemination rate, kg/sec

δ_v = dielectric constant of medium surrounding particle, dimensionless

$\underline{\epsilon}$ = permittivity of free space = 8.854×10^{-12} $(coulomb)^2/(m)^2(newton)$

μ = absolute gas viscosity, (kg)/(m)(sec) or dekapoise

ρ_p = particle density, kg/m^3

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11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
Dissemination investigations of liquid and solid agents	Edgewood Arsenal Research Laboratories Edgewood Arsenal, Maryland 21010

13. ABSTRACT

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CS						
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Laboratory nozzle						
Hydropholic powders						
Electrically charged particles						